

# PATENT ABSTRACTS OF JAPAN

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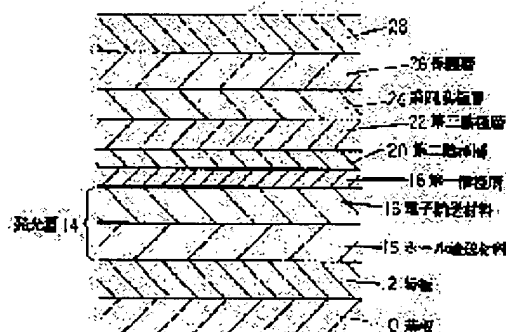
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## (54) ELECTROLUMINESCENT ELEMENT AND MANUFACTURE THEREOF

(57)Abstract:

PROBLEM TO BE SOLVED: To simplify the structure, to improve the light emitting efficiency, to prolong the lifetime, and to enable the even light emission without generating a dark spot.

SOLUTION: A positive electrode 12 made of the transparent electrode material such as ITO is formed on a transparent board 10 such as glass or the like, and EL material such as a hole transporting material 15 and an electron transporting material 16 are laminated on the top surface of the transparent positive electrode 12, and a thin layer of a first negative electrode layer 18 of a relatively high purity is laminated on the surface of the EL material, and furthermore, a thin layer of a second negative electrode layer 20 of a relatively low purity, which has high electron donative characteristic, is laminated thereon. A third negative electrode layer 22 of a relatively high purity is formed on the surface of the second negative electrode layer 20 at a thickness larger than that of the first negative electrode layer 18, and a fourth negative electrode layer 24 having electron donative characteristic higher than that of the second negative electrode layer 20 is formed on the surface of the third negative electrode layer 22 at a thickness larger than that of the second negative electrode layer 20. A protecting layer for shielding the fourth negative electrode layer 24, which is made of metal thin film or resin, from outside air is formed on the fourth negative electrode layer 24.



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CLAIMS

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[Claim(s)]

[Claim 1] An anode plate is formed of a transparent electrode material on a transparent substrate, and the laminating of the EL material of hole transportation material and electronic transportation material is carried out to the upper surface of the transparent anode plate. The laminating of the first catholyte of a high grade is relatively carried out to the front face of the above-mentioned EL material thinly. Although purity is still more relatively [ front face / the ] low, the laminating of the second electron-donative high catholyte is carried out thinly. The EL element by which the third catholyte of a high grade was formed relatively [ front face / of this second catholyte ] more thickly than the first catholyte of the above, and the fourth high catholyte / thicker than the second catholyte of the above and / more nearly electron-donative than the second catholyte of the above was formed in the front face of this third catholyte.

[Claim 2] The above-mentioned EL material is an EL element according to claim 1 which is organic EL material.

[Claim 3] The EL element according to claim 1 or 2 in which the protective layer which intercepts this fourth catholyte with the open air was formed on the fourth catholyte of the above.

[Claim 4] On a transparent substrate, the anode plate of a transparent electrode material is formed by the vacuum thin film coating technology. The laminating of the EL material is carried out to the upper surface of the transparent anode plate by the above-mentioned vacuum thin film coating technology. Furthermore, the laminating of the first catholyte of a high grade is relatively carried out to the front face of the above-mentioned EL material thinly by the above-mentioned vacuum thin film coating technology. Although purity is still more relatively [ front face / the ] low, the laminating of the second electron-donative high catholyte is thinly carried out by the above-mentioned vacuum thin film coating technology. The laminating of the third catholyte of a high grade is carried out by the above-mentioned vacuum thin film coating technology relatively [ front face / of this second catholyte ] more thickly than the first catholyte of the above. The manufacture method of the EL element which carries out the laminating of the fourth high catholyte [ thicker than the second catholyte of the above and ] more nearly electron-donative than the second catholyte of the above to the front face of this third catholyte by the above-mentioned vacuum thin film coating technology.

[Claim 5] The manufacture method of the EL element according to claim 4 which forms the protective layer which intercepts this fourth catholyte with the open air by the above-mentioned vacuum thin film coating technology on the fourth catholyte of the above, and applies the protective layer of a resin to the front face.

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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] This invention relates to the EL element used for the flat-surface light source or a display, and its manufacture method.

[0002]

[Description of the Prior Art] As shown in drawing 2, a conventional, for example, organic, EL (electroluminescence) element forms the anode plate 2 of the ITO film of a translucency in a glass substrate 1, forms the hole transportation material 3, such as a triphenylamine derivative (TPD), in the upper surface, and is carrying out the laminating of the electronic transportation material 4, such as an aluminum chelate complex (Alq3), on it. And the cathode 5, such as aluminum, Li, Ag, Mg, and In, is formed in the upper surface. Predetermined voltage is impressed between an anode plate 2 and cathode 5, and this organic EL element produces luminescence. And on the occasion of manufacture of this organic EL element, an electrode material and EL material are formed with vacuum deposition on a glass substrate 1.

[0003]

[Problem(s) to be Solved by the Invention] Here, it is desired for the electronic supply nature by which cathode 5 supplies an electron to the electronic transportation material 4 to be high, and it is required that those of the material with a work function and this number of \*\*\*\*\* should be low as the index. Although there are Na, Li, Mg, calcium, etc. as a low material of this work function, a material usable to an EL element is restricted to Li, Mg, etc. from the ease of handling.

[0004] however, generally the thing of a high grade was hard to be obtained highly, and electron-donative high material, i.e., the low material of a work function, such as Li, Mg, etc. which are the material which constitutes the cathode 5 of the above-mentioned Prior art, had the same reactivity also in those alloys, and the purity which is usually about 99% was a limitation. And existence of an impurity caused a dark spot of an EL element, and the case where dark spot's existence area reached 30% of a luminescence side had become \*\* and a practical use top problem at about 99% of purity. On the other hand, although the thing of a high grade was obtained, material, such as aluminum other than these, was inferior in electronic supply nature, and had the problem that the luminous efficiency and the life of an EL element fell.

[0005] Moreover, the EL element which carried out the laminating of the second catholyte by the electron-donative high aluminum-Li alloy by the applicant for this patent although the laminating of the first catholyte, such as aluminum of a high grade, was relatively carried out to the front face of EL material and purity was still more relatively [ front face / the ] low in order to solve the above-mentioned trouble is also proposed. However, when the amount of Li becomes there were less, luminous efficiency fell, when aluminum layer of a high grade was thickened, and aluminum layer was made thin also according to this structure, the influence of

an impurity became strong, and there was an opposite problem of becoming easy to generate a dark spot. [ few ]

[0006] This invention was made in view of the above-mentioned Prior art, and it is easy composition, its luminous efficiency is good, and its life is long, and it aims at offering the EL element which enables uniform luminescence without a dark spot, and its manufacture method.

[0007]

[Means for Solving the Problem] As for this invention, the anode plate by transparent electrode materials, such as ITO, is formed on transparent substrates, such as glass. The laminating of the EL material of hole transportation material and electronic transportation material is carried out to the upper surface of the anode plate of the transparent electrode. Furthermore, the laminating of the first catholyte of a high grade is relatively carried out to the front face of the above-mentioned EL material thinly. Although purity is still more relatively [ front face / the ] low, the laminating of the second electron-donative high catholyte is carried out thinly. It is the EL element by which the third catholyte of a high grade was formed relatively [ front face / of this second catholyte ] more thickly than the first catholyte of the above, and the fourth high catholyte / thicker than the second catholyte of the above and / more nearly electron-donative than the second catholyte of the above was formed in the front face of this third catholyte. Furthermore, the protective layer which intercepts this fourth catholyte with the open air is formed with a metal thin film or a resin on the fourth catholyte of the above.

[0008] This invention on transparent substrates, such as glass, moreover, by vacuum deposition, sputtering, and other vacuum thin film coating technologies Form the anode plate of transparent electrode materials, such as ITO, and the laminating of the EL material of hole transportation material and electronic transportation material is carried out to the upper surface of the anode plate of the transparent electrode by the above-mentioned vacuum thin film coating technology. Furthermore, the laminating of the first catholyte of a high grade is relatively carried out to the front face of the above-mentioned EL material thinly by the above-mentioned vacuum thin film coating technology. Although purity is still more relatively [ front face / the ] low, the laminating of the second electron-donative high catholyte is thinly carried out by the above-mentioned vacuum thin film coating technology. The laminating of the third catholyte of a high grade is carried out by the above-mentioned vacuum thin film coating technology relatively [ front face / of this second catholyte ] more thickly than the first catholyte of the above. It is the manufacture method of the EL element which carries out the laminating of the fourth high catholyte [ thicker than the second catholyte of the above and ] more nearly electron-donative than the second catholyte of the above to the front face of this third catholyte by the above-mentioned vacuum thin film coating technology. Formation of each class by the above-mentioned vacuum thin film coating technology is performed at a series of processes. Furthermore, the protective layer which intercepts this fourth catholyte with the open air is formed by the above-mentioned vacuum thin film coating technology on the fourth catholyte of the above, and the protective layer of a resin is applied to the front face.

[0009] The EL element of this invention forms the thin film of the first catholyte with purity high as cathode, and the second electron-donative high catholyte. by this In order for there to be almost no dark spot, to make it luminous efficiency also become good by the second catholyte and to improve luminous efficiency further more Electronic supply nature prepares the fourth higher catholyte through the third catholyte of a high grade, electron-donative high metal atoms, such as Li, are spread in the third catholyte, there is no dark spot further, luminous efficiency can also be good and a life can also make it long.

[0010]

[Embodiments of the Invention] Hereafter, the gestalt of implementation of this invention is explained based on a drawing. Drawing 1 shows 1 operation gestalt of the EL element of this

invention, and the EL element of this operation gestalt is an organic thin film EL element, and the anode plate 12 which are transparent electrodes, such as ITO, is formed in the front face of the transparent substrates 10, such as glass, a transparent resin, and a quartz, so that it may illustrate. The luminous layer 14 by EL material is formed in the front face of an anode plate 12. And it is formed in the thickness whose first catholyte 18 by aluminum of 99.999% or more of purity is about 50A in the front face of a luminous layer 14, and Li is formed in the upper surface at the thickness whose second cathode 20 of 0.01% or less of minute amount \*\*\*\* aluminum-Li alloy is about 50A. The rate of Li of the second catholyte 20 was made into 0.01% or less by lessening Li for lessening an impurity and making it an alloy with purity high as much as possible.

[0011] On the second catholyte 22, it is formed in the thickness whose third catholyte 22 by aluminum of 99.999% or more of purity is about 200A, and the fourth cathode 24 of the aluminum-Li alloy which contains Li about 0.05% on the upper surface is formed at the thickness which is about 200A. The rate of Li of the fourth catholyte 24 was made into about 0.05% for making [ more ] Li than the second catholyte 20, making [ many ] the diffusing capacity to the third catholyte 22 of Li, and making luminous efficiency higher. In addition, what is necessary is to be able to set up the thickness of the second catholyte 18 and 20 suitably, and it to be relatively thinner than the third and the fourth catholyte 22 and 24, and just to make it into what has high purity for a start. And \*\*\*\*\* proper change of thickness of 100 to about 1000A is respectively possible for the third and the fourth catholyte 22 and 24, and electronic supply nature should just make them more preferably the thickness of 200 to about 500A as a range which a dark spot cannot generate easily well.

[0012] The luminous layer 14 of EL material has a triphenylamine derivative (TPD), a hydrazone derivative, an arylamine derivative, etc. as a hole transportation material 15 among parent material. Moreover, as an electronic transportation material 16, an aluminum chelate complex (Alq3), a distyrylbiphenyl derivative (DPVBi), an OKISA diazole derivative, a screw CHIRIRU anthracene derivative, a benzo oxazole thiophene derivative, perylenes, and thiazoles are used. The ratio of the above-mentioned hole transportation material 15 and the electronic transportation material 16 can be suitably changed in 10:90 or 90:10.

[0013] The laminating of the protective layer 26 is carried out to the upper surface of the fourth catholyte 24. A protective layer 26 is formed of metal thin films, such as Ag and aluminum, and intercepts each catholytes 18, 20, 22, and 24 and a luminous layer 14 from the open air. And a protective layer 28 is formed by resins, such as a phenol and epoxy, and the conductive paint.

[0014] The manufacture method of the organic thin film EL element of this operation gestalt forms the anode plate 12 by ITO etc. on a substrate 10 first at the whole surface by the thin film coating technology in the vacuum of vacuum evaporatio, flash plate vacuum evaporatio, sputtering, and others. Next, the luminous layer 14 of EL material is formed one by one by the arbitrary methods of the above-mentioned thin film coating technology in order of the hole transportation material 15 and the electronic transportation material 16. The layer of the conductive material 15 is formed in the front face of the layer of the EL material 14 at the whole surface by the arbitrary methods of the above-mentioned vacuum thin film coating technologies. Then, the first catholyte 18 is formed in thickness predetermined by the above-mentioned vacuum thin film coating technologies, such as vacuum evaporatio. Next, the second catholyte 20 is formed similarly.

[0015] Next, the third catholyte 22 is similarly formed in the front face of the second catholyte 20, and the fourth catholyte 24 is further formed in it similarly. And a protective layer 26 is formed. A protective layer 26 forms aluminum and Ag similarly by the above-mentioned vacuum thin film coating technology. Even formation of this protective layer 26 is performed to a series in a vacuum furnace. And except for the takeoff connection of an electrode, you may form the protective layer 28 by the resin by application further. In addition, a protective layer 26

has desirable Ag, when soldering nature is taken into consideration.

[0016] A degree of vacuum is  $6 \times 10^{-6}$  Torr, and, in the case of EL material, forms vacuum evaporation conditions by the evaporation rate of 50A/sec here. A flash plate vacuum deposition drops the source of vacuum evaporation which heated preferably 300-600 degrees C of organic EL material beforehand mixed by the predetermined ratio at 400-500 degrees C, and evaporates organic EL material at a stretch. Moreover, the organic EL material is held into a container, the container is heated quickly, and vacuum evaporation may be carried out at a stretch.

[0017] The EL element of this operation gestalt has secured a certain amount of luminous efficiency, forming the first catholyte 18 of aluminum with purity high as cathode, purity forming thinly the second high catholyte 20 of an AL-Li alloy also with high electronic supply nature, there being no impurity, and electronic supply nature also forming to some extent high cathode, and pressing down generating of a dark spot. And in order to form the third catholyte 14 of a high grade thickly relatively, to suppress the influence of an impurity further and to raise electronic supply nature more, the fourth catholyte 26 of an AL-Li alloy with more rates of Li than the second electrode layer is formed. It is spread in large quantities [ Li ] in the third catholyte by there being no dark spot at the time of luminescence by the first catholyte 18 high [ of purity ] by this, and the second catholyte 20 comparatively high [ of purity ], and luminous efficiency's also carrying out right not-less reservation to some extent, and forming the fourth catholyte 26 with much Li through the third catholyte 22 still higher [ of purity ], the electronic supply nature of the third catholyte 24 is raised, and luminous efficiency becomes better.

[0018] In addition, Ag and In which a high grade is obtained in addition to aluminum of a high grade, and do not have a bad influence on a luminous layer are sufficient as the third catholyte for a start [ of the EL element of this invention ]. Moreover, the second and the fourth catholyte can change the component ratio of an Al-Li alloy suitably, and alloys, such as other Mg, and a high metallic-material simple substance electron-donative in addition to this are sufficient as them. Furthermore, efficiency can be further gathered by keeping temperature at about 100 degrees C after forming the second and the fourth catholyte, and promoting diffusion.

Moreover, EL material is also selectable things, such as what mixed and deposited others and these, suitably, although the laminating of hole transportation material and the electronic transportation material was carried out, and the luminescent color is also arbitrarily selectable.

[0019]

[Effect of the Invention] The EL element and its manufacture method of this invention carry out the laminating of the material with high purity to a luminous layer thinly as cathode. Carry out the laminating of the high material electron-donative on it thinly, and the laminating of the cathode material of a high grade is further carried out thickly relatively on it. Since a high material electron-donative moreover was formed thickly relatively, there is no dark spot, moreover, luminous efficiency is good and a life can also make an EL element long.

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TECHNICAL FIELD

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[The technical field to which invention belongs] This invention relates to the EL element used for the flat-surface light source or a display, and its manufacture method.



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DESCRIPTION OF DRAWINGS

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[Brief Description of the Drawings]

[Drawing 1] It is the cross section of the EL element of 1 operation gestalt of this invention.

[Drawing 2] It is the cross section of the EL element of \*\*\*\*\*.

[Description of Notations]

- 10 Substrate
- 12 Anode Plate
- 14 Luminous Layer
- 15 Hole Transportation Material
- 16 Electronic Transportation Material
- 18 First Catholyte
- 20 Second Catholyte
- 22 Third Catholyte
- 24 Fourth Catholyte
- 26 28 Protective layer

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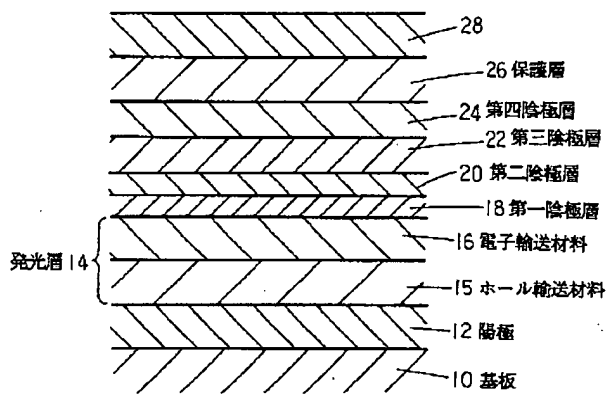
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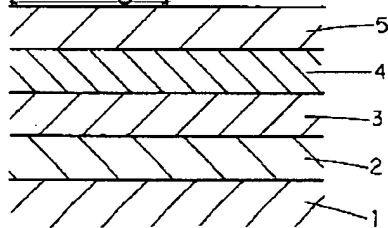
DRAWINGS

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[Drawing 1]



[Drawing 2]




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